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Abstract

With the increasingly wide spread use of catalytic converters for meeting exhaust emission regulations considerably attention is currently being directed towards improving their performance. An attractive alternative to experimental analysis is computational modelling. The paper shows that the equations that govern the behaviour of catalysts can be solved using a commercial CFD code.

1 Introduction

The increasing awareness of the impact that automotive exhaust emissions have on the environment has lead, in many countries, to the adoption of emissions regulations. Most of these regulations, which are becoming ever more strict, dictate that at present exhaust catalytic converters are the only practical solution to the emissions problem. To limit the impact catalysts have on engine performance and fuel consumption considerable research is being undertaken to find ways of improving the performance of catalysts so that their size can be reduced. The smaller their size the smaller the impact on engine performance. Reducing size also brings benefits with component cost and packaging considerations. Reduced size however needs to be achieved without prejudicing component longevity or cold start characteristics, known as catalyst light-off.

Before new improved catalyst designs can be built an understanding of the factors and parameters that affect performance must be obtained. There are in essence two approaches to this; experimental analysis or computational modelling. This paper outlines the general equations that govern the behaviour of automotive exhaust catalysts and presents a method of incorporating them into a commercial computational fluid dynamics (CFD) code such that fundamental trends in behaviour can be studied.

2 Factors Affecting Catalyst Performance

The salient factors that are likely to affect catalyst performance are included in Figure 1. Although they have been recognised for sometime the relative importance of each parameter is still to be fully understood. To gain a better understanding many parametric studies need to be carried out both experimentally and computationally.

The type of catalyst reactor under consideration is the monolith type. These consist of a series of several thousand parallel capillary passages, typically approximately 1 mm in diameter, running axially through a metallic or ceramic substrate. Note that because the exhaust gas flow profile across the front face of the monolith is highly non-uniform, as a result of using a short inlet diffuser, the distribution of fluid properties throughout the system is three-dimensional in nature.

3 Computational Modelling

Potentially computational modelling has a number of advantages over experimental analysis. Designing and building test rigs, test components and prototypes is costly and time consuming. However if an

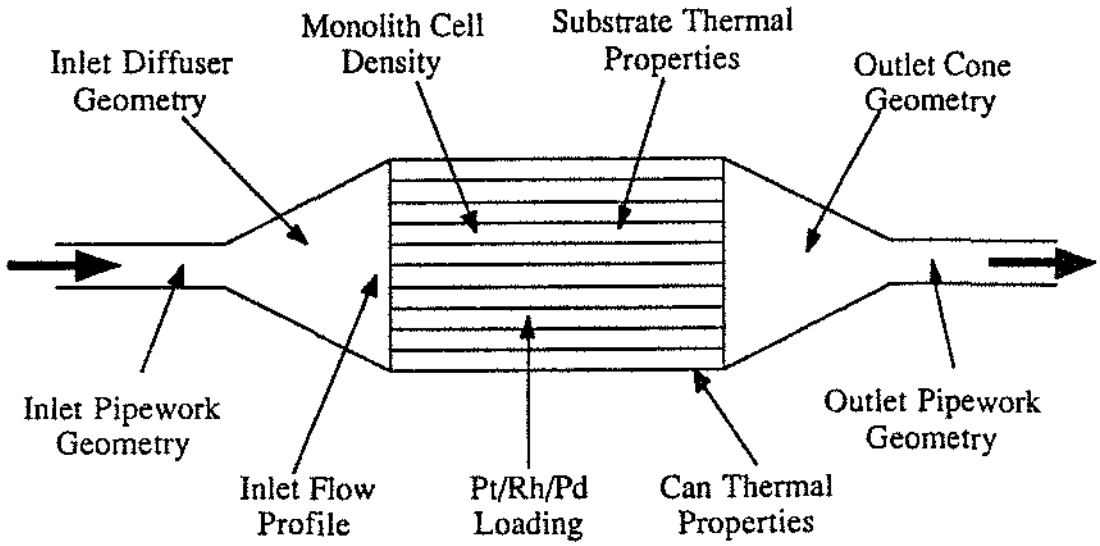

Figure 1 - Factors Affecting Catalyst Performance

accurate, reliable and computationally fast computer model is available considerable savings on time and money can be made. Ultimately a computational model that can simulate catalyst behaviour fully is desired such that performance of potential designs can be tested before any prototypes are made. However at present a full model is impractical. Because of the non-linear nature of the governing equations a numerical procedure of the finite volume type is required to obtain solutions. It has been shown [1] that to numerically model every channel of a monolith reactor a computational grid of the order of 10^7 cells would be required. To model the pulsed flow that exists in automobile exhausts a transient analysis using time steps of 0.001s duration is needed. The speed of most existing computers puts such simulations outside the capabilities of most organisations. Another factor that prevents full modelling of catalysts is the fact that a full mathematical representation of the chemical kinetics occurring in the catalyst has yet to be achieved.

Although a full computational model is presently impractical somewhat simplified transient models have been developed, notably by Flytzani-Stephanopoulas et al. [2], Zygourakis [3] and Chen et al. [4]. All these models use an approach known as an equivalent continuum technique. With such a technique the bulk behaviour of the dependent variables is modelled. Unfortunately all these existing models suffer from a significant weaknesses; they are all unable to simulate the feedback effect the chemical reactions have on the velocity distribution across the monolith face. To do this the fluid dynamics occurring upstream of the catalyst in the inlet ductwork and diffuser have to be included in the model.

4 Governing Equations

The equations that predict the thermofluid dynamics and chemistry that occur in catalysts have been established for sometime. The equations given below are appropriate for an equivalent continuum approach, being slightly modified forms of the general transport and conservation equations. They are essentially those described by Zygourakis [3] with extensions to include the behaviour of the exhaust gas outside the catalyst monolith and rearranged into a form in which they can be implemented within a CFD code.

For the fluid dynamics that occur in the inlet pipework, diffuser and outlet cone the Reynolds averaged Navier-Stokes equations are solved. Closure is provided by use of one of the standard turbulence models. The flow in the monolith substrate is assumed to be unidirectional and laminar with the cross sectional shape of each channel being taken as circular. Neglecting entrance effects the governing equation for such flow is give by the Hagen-Poiseuille equation,

$$\frac{\partial p}{\partial x} = -\frac{32\nu\rho u}{b^2} \tag{1}$$

The temperature of the exhaust gas, T_g, is obtained by solving the conservation equation for the gas enthalpy,

$$\frac{\partial \rho_g h_g}{\partial t} + \nabla.\left(\rho_g U h_g\right) - \nabla.\left(\frac{k_g}{c_{pg}}\nabla h_g\right) = S1 \tag{2}$$

The source term S1 in EQ (2) is only active in the part of the calculation domain representing the catalyst. It is given by,

$$S1 = \frac{h a_v}{c_{pg}}\left(T_s c_{pg} - h_g\right) \tag{3}$$

The conservation equations for the chemical species in the exhaust gas are of the form,

$$\frac{\partial \rho_g C_{gi}}{\partial t} + \nabla.\left(\rho_g U C_{gi}\right) - \nabla.\left(\rho_g D_i \nabla C_{gi}\right) = S2 \tag{4}$$

where C_{gi} is the mole fraction of the species. The source term S2 in EQ (5), like S1 in EQ (2), is only active in the catalyst. It is given by,

$$S2 = \rho_g K_{mi} a_v (C_{si} - C_{gi}) \tag{5}$$

The concentrations of such species on the monolith surface, where the reactions take place, are governed by expressions of the form,

$$v_c \frac{\partial C_{si}}{\partial t} = K_{mi} a_v \left(C_{gi} - C_{si}\right) - \frac{M_{Rg}}{10^3 \rho_g} a_c R_i \tag{6}$$

Note that the transient term in EQ (7) is frequently neglected an assumption that is justified on the grounds that reaction rates occur so quickly they can be considered instantaneous. The reaction rate expressions R_i are of the Arrhenius equation type.

The variable that links all the others together is the temperature of the substrate T_s. It is the only variable that can be transported between adjacent monolith channels. The equation governing its behaviour is essentially that of heat conduction in a solid. However, because an equivalent continuum approach is being used the orthotropic nature of the heat conduction in a monolith must be taken into consideration. Thus the governing equation becomes,

$$\rho_s \frac{\partial T_s}{\partial t} - \frac{k_s}{c_{ps}} \left\{ \frac{\partial^2 T_s}{\partial x^2} + \frac{G}{(1-\varepsilon)} \left(\frac{\partial^2 T_s}{\partial y^2} + \frac{\partial^2 T_s}{\partial z^2} \right) \right\} = S3 + S4 \qquad (7)$$

Note that x is the flow direction of the gas, whereas y and z are the Cartesian directions perpendicular to the flow. G is defined as k_{eff}/k_s, where k_{eff} is the effective thermal conductivity of the composite of exhaust gas and substrate in the direction of y and z. The two source terms S3 and S4 are given by,

$$S3 = \frac{h a_v}{(1-\varepsilon) c_{ps}} \left(T_g - T_s \right) \qquad (8)$$

$$S4 = \frac{a_c R_i \Delta H_i}{(1-\varepsilon) c_{ps}} \qquad (9)$$

Several auxiliary relationships need to be used such as calculating the gas density from the ideal gas law and making viscosity a function of temperature. The heat and mass transfer coefficients, h and K_{mi}, are calculated from Nusselt and Sherwood numbers respectively. For simplicity these can be assumed constant throughout the monolith. The boundary conditions used for the above equations vary from case to case.

5 Equation Solutions

Although it would be feasible to write a purpose built computer code to solve the above equations a considerable amount of development time would be needed as well as a detailed knowledge of numerical techniques. An attractive alternative is to utilise the solution algorithms of a general purpose commercial CFD code. The CFD code selected to solve the equations in the present study was PHOENICS, supplied by CHAM Ltd.

Table 1

CATALYST PARAMETERS

Monolith Dimensions:		Material Properties:	
L	152.0 mm	ρ_s	1537 kg/m^3
r	46.48 mm	ρ_m	0.13 kg/m^3
b	1.017 mm	c_{ps}	1000 J/kgK
ε	0.6408	c_{pm}	600 J/kg K
a_v	1981 m^2/m^3	k_s	1.476 W/m K
a_c	537.9x10^4 m^{2}Pt/m^3	G	0.1795
Inlet Conditions:		M_{Rg}	29 kg/kmol
C_{gCO}	0.02 mol/mol	c_{pg}	1089 J/kg K
C_{gNO}	5.0x10^{-4} mol/mol	Pr_g	0.7
$C_{gC_3H_6}$	4.5x10^{-4} mol/mol	D_{CO}	1.332x10^{-4} m^2/s
C_{gO_2}	0.05 mol/mol	K_{mCO}	0.4793 m/s
$\dot{m}$	29.2 g/s		

To illustrate the capabilities of the model a selection of results have been taken from a simulation of a typical catalyst geometry. A listing of the catalyst dimensions and material properties is included as

Table 1. The results presented are for a transient cold start simulation. The incoming exhaust gas has an exponential temperature rise given by the expression,

$$T_{g(Inlet)} = 200\left(3 - e^{-t/37.74}\right) \qquad (10)$$

Heat loss to the external environment is included through the catalyst insulating mat, the external air temperature being set at 300K. The oxidation of CO is taken as a single representative chemical reaction. Figure 2 gives the CO conversion efficiency against time. Figures 3 and 4 present contour plots of T_s and T_g as they evolve with time. As can be seen the catalyst lights-off at approximately 40 seconds.

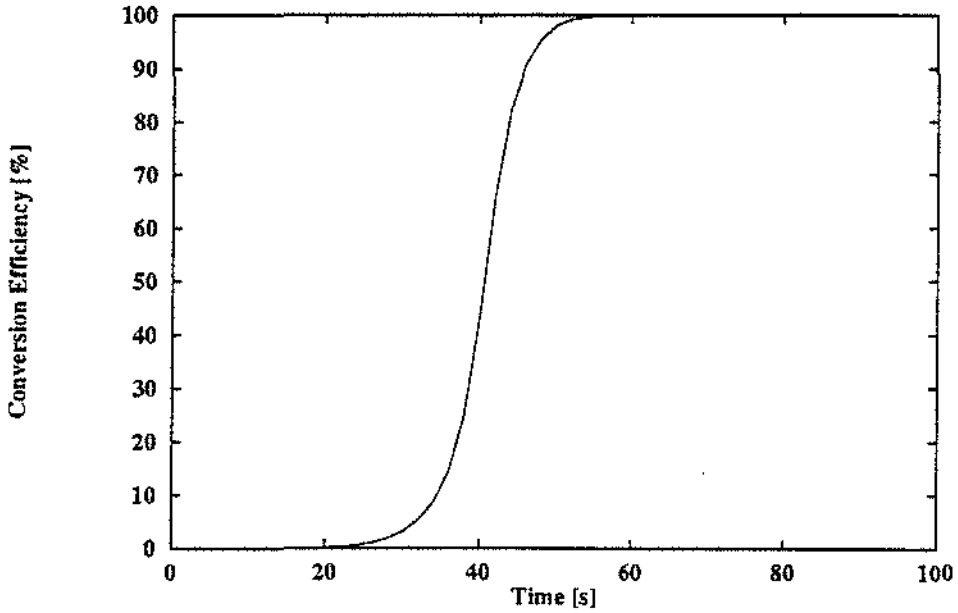

Figure 2 - CO Conversion Efficiency

6 Conclusions

This paper shows that the equations that govern the behaviour of catalysts can be solved using a commercial CFD code. Because of the uncertainties over the values of some of the coefficients in the mathematical expressions the results presented cannot be used to accurately validate the model against experimental data. However a light-off time of 40 seconds is typical for catalysts of the type being considered, under the conditions being modelled. The other effects illustrated by the results are also typical of known catalyst behaviour. Thus it is reasonable to conclude that the model presented here can be used to study important catalyst phenomena.

7 Nomenclature

a_c	- catalyst area per unit reactor volume, m^2Pt/m^3
a_v	- ratio of reactor surface to reactor volume, m^2/m^3
b	- hydraulic diameter of monolith channels, m
c_{pg}	- specific heat of exhaust gas, J/kg K
c_{ps}	- specific heat of monolith material, J/kg K
c_{pm}	- specific heat of insulating mat, J/kg K
C_{gi}	- concentration of gas species i, mol/mol

c_{pm} - specific heat of insulating mat, J/kg K
C_{gi} - concentration of gas species i, mol/mol
C_{si} - concentration of species i on monolith surface, mol/mol
D_i - diffusivity of species i, m^2/s
G - monolith orthotropic conductivity factor
h - heat transfer coefficient, W/m^2 K
ΔH_i - heat of reaction of species i, J/mol
h_g - enthalpy of exhaust gas, J/kg
k_g - thermal conductivity of exhaust gas, W/m K
k_s - thermal conductivity of monolith material, W/m K
K_{mi} - mass transfer coefficient for species i, m/s
L - length of monolith, m
$\dot{m}$ - exhaust mass flow rate, kg/s
M_{Rg} - relative molecular mass of exhaust gas, kg/kmol
p - pressure, N/m^2
Pr_g - Prandtl number of exhaust gas, $c_{pg}\nu\rho_g/k_g$
r - radius of monolith, m
R_i - reaction rate of species i, $mol/m^2Pt\ s$
t - time, s
T_g - temperature of exhaust gas, K
T_s - temperature of monolith, K
u - axial velocity in monolith, m/s
U - velocity of exhaust gas, m/s
v_c - volume available to store chemical species per unit reactor volume, m^2/m^2
x,y,z - co-ordinate axes
ε - monolith porosity
ν - kinematic viscosity, m^2/s
ρ_g - density of exhaust gas, kg/m^3
ρ_s - density of monolith material, kg/m^3
ρ_m - density of insulating mat, kg/m^3

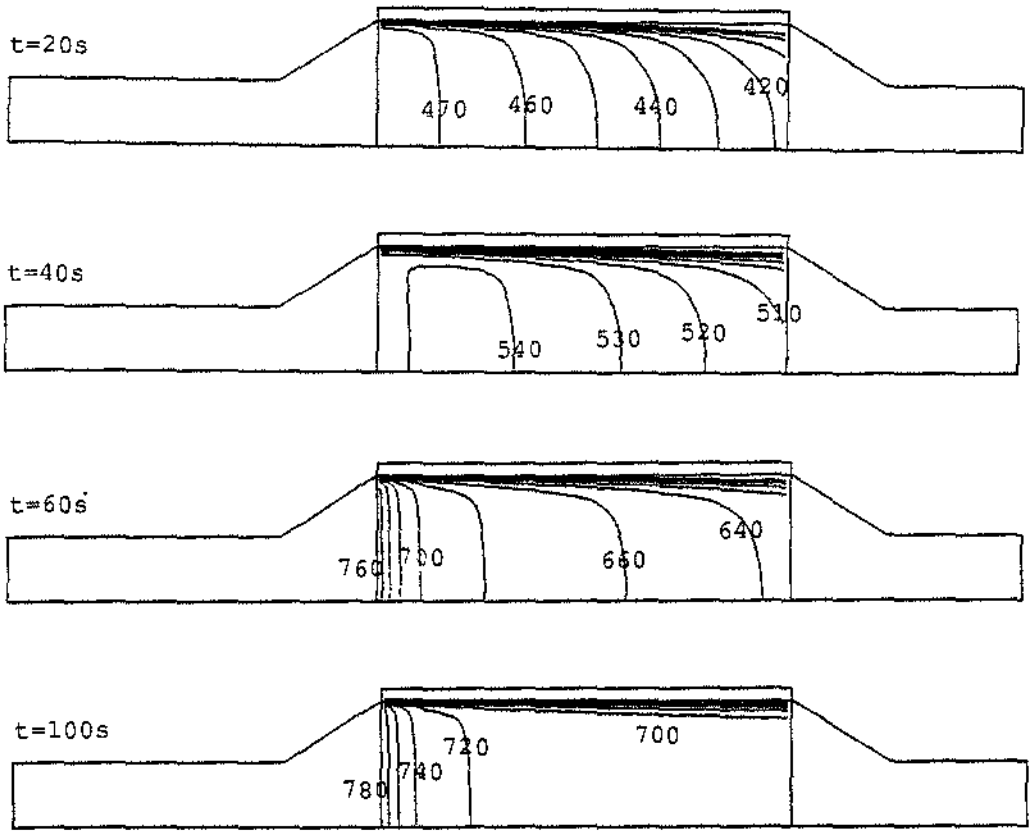

Figure 3 - Evolution of substrate temperature (in K)

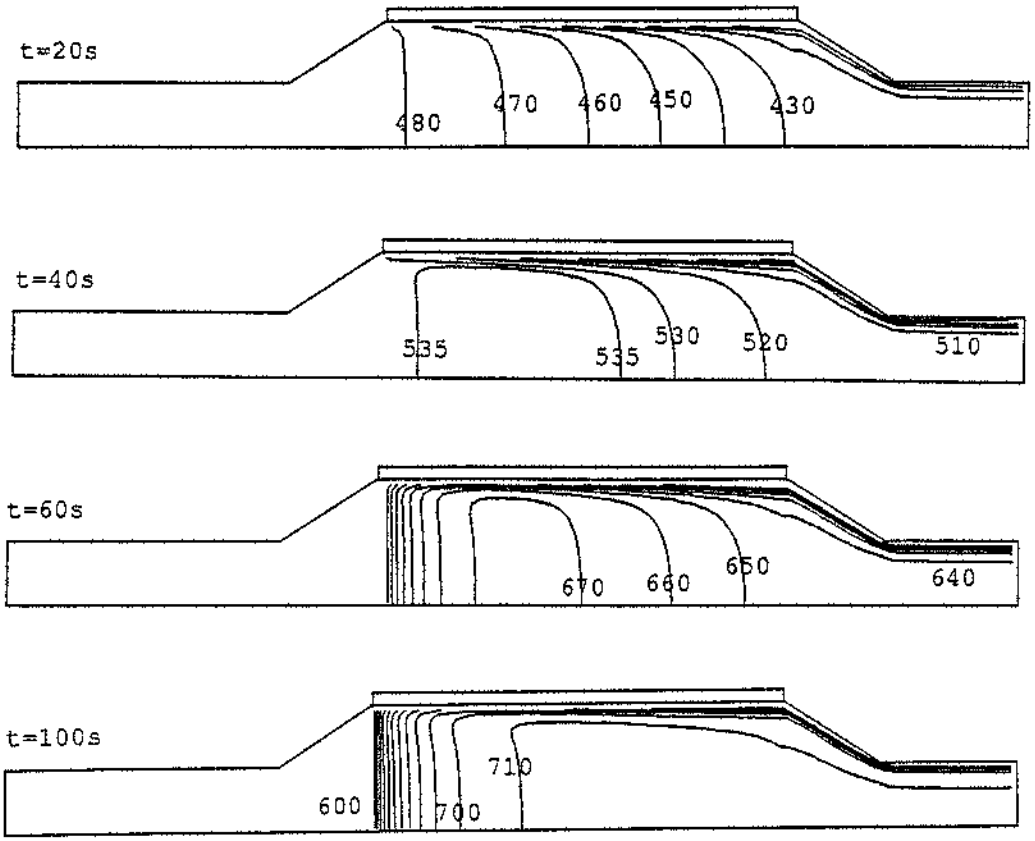

Figure 4 - Evolution of gas temperatures (in K)

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